Research paper

Use of lead isotopes for developing chronologies in recent salt-marsh sediments

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ABSTRACT

Dating of recent salt-marsh sediments is hindered by the radiocarbon plateau and the moving ~100 year window of ^{210}Pb accumulation histories. Introduction of anthropogenic Pb to the environment is a means to date salt-marsh sediment deposited over the last 200 years by correlating downcore changes in concentration and isotopic ratios to historical production and consumption. We investigated use of Pb as a chronometer in a core of salt-marsh sediment from New Jersey, USA. Changes in Pb concentration identified horizons at AD 1875, 1925, 1935 and 1974 that correspond to features of historic U.S Pb production and consumption. Stable lead isotopes (^{206}Pb:^{207}Pb) constrained ages at AD 1827, 1857 and 1880, reflecting Pb production in the Upper Mississippi Valley with its unusual isotopic signature and at AD 1965 and 1980 from leaded gasoline. Confirmatory evidence for the gasoline horizons came from increased Sb concentrations caused by vehicle emissions and industrial activity. These nine chronostratigraphic markers of fixed dates provide precise constraints on sediment age in the northeastern USA and enable salt-marsh records of coastal evolution and wetland development to extend beyond the period of instrumental measurements.

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1. Introduction

Salt marshes accrete vertically through vegetative growth and detrital sedimentation of organic and mineral particles (Chapman, 1960; Orson et al., 1998). Material is also deposited directly from the atmosphere, including Pb (and other elements) emitted by industrial processes. The resulting sedimentary sequences can form continuous accumulations of salt-marsh peat that are well suited to reconstructing wetland development, environmental change and sea level, if accurate and precise chronologies are developed. Abundant organic material (including plant macrofossils) has made radiocarbon (^{14}C) dating the principal means of determining the age of salt-marsh sediment (Törnqvist et al., 1992, 2004; van de Plassche et al., 1998; Shennan and Horton, 2002; Engelhart et al., 2011). Difficulties with this technique hinder its application to sediments younger than about 350 years. Between AD 1650 and 1950, a plateau in the ^{14}C calibration curve caused by variability in atmospheric ^{14}C production hampers chronological interpretation by producing multiple calendar ages for a single sample (Reimer and Reimer, 2007). Since AD 1950 introduction of anthropogenic ^{14}C from above-ground testing of nuclear weapons has significantly altered atmospheric ^{14}C concentration (Shotyk et al., 2003; Hua and Barbetti, 2004; McGee et al., 2004). Although new approaches to ^{14}C dating have increased its usefulness for estimating the age of recent sediments (Marshall et al., 2007; Hua, 2009), studies seeking to provide detailed histories of recent salt-marsh accretion rely upon alternative techniques to achieve the desired temporal resolution for the last 350 years.

The most widely used radionuclide for estimating the accumulation rate and relative age of recent salt-marsh sediments is ^{210}Pb (Gehrels et al., 2005; Kemp et al., 2009). Use of ^{210}Pb as a sediment chronometer involves assumptions regarding how ^{210}Pb is delivered from the atmosphere to sediments (Robbins, 1978; Appleby and Oldfield, 1992; Appleby, 2001). The relative contribution of atmospheric and tide-water ^{210}Pb sources to salt marshes cannot be determined with certainty, therefore sediment age estimates from ^{210}Pb are corroborated by independent means.

Radiocesium (^{137}Cs) is most frequently used in this capacity as it provides an absolute time marker for peak fallout in AD 1963–64 (Ritchie and McHenry, 1990). A limitation of ^{210}Pb geochronology is that it is restricted to the last 100–120 years, and this range moves forward each year leaving a growing "*chronological vacuum*" (Gale, 2009).

Stable Pb isotopes can extend sediment chronologies developed using ^{210}Pb and ^{137}Cs, and narrow the crucial gap (approximately AD 1650–1900) between time scales covered by these radionuclides and ^{14}C geochronology. The age and geological history of mineral deposits results in Pb ores with differing isotopic compositions (Russell and Farquhar, 1960; Chow and Earl, 1972). During industrial processes (including production and consumption of leaded gasoline), there is minimal fractionation of Pb isotopes and emissions preserve the Pb isotopic signature of the ore from which they were derived (Ault et al., 1970; Doe, 1970). Prior to AD 1979 as much as 1.96×10^{10} kg of Pb was added to the atmosphere from global anthropogenic emissions (Nriagu, 1979), primarily high-temperature industrial activities and combustion of leaded gasoline (Kelly et al., 2009). The atmospheric residence time of Pb aerosols permits their dispersal and subsequent deposition over continental-scale distances in directions controlled by prevailing winds (Sturges and Barrie, 1987; Wu and Boyle, 1997; Bollhöfer and Rosman, 2000, 2001), as shown by measurements at sites distal to source regions, such as Greenland (Rosman et al., 1993; Hong et al., 1994), Bermuda (Shen and Boyle, 1987; Kelly et al., 2009), and deep ocean basins (Schaule and Patterson, 1981; Shen and Boyle, 1987; Hamelin et al., 1997; Wu et al., 2010). There is close agreement between the timing of emissions and deposition (Graney et al., 1995). As the variability of source material is greater than the precision of analytical measurements (Shen and Boyle, 1987; Chillrud et al., 2003), ratios of stable Pb isotopes preserved in sediment can distinguish among different sources of Pb (Shotyk et al., 1998; Kamenov et al., 2009). Most investigations of historical Pb deposition used independent chronological control or correlation with dated sedimentary records to describe the timing of Pb contamination and to apportion sources of pollution (Edgington and Robbins, 1976; Graney et al., 1995; Cochran et al., 1998; Marcantonio et al., 2002; Lima et al., 2005a; Kamenov et al., 2009). Alternatively, changing concentrations and ratios of Pb isotopes measured in sediments can be ascribed to historical variations in Pb production and consumption with well-established ages (Shen and Boyle, 1987; Graney et al., 1995; Farmer et al., 2001; Marcantonio et al., 2002; Chillrud et al., 2004; Lima et al., 2005a; Gehrels et al., 2008; Kelly et al., 2009; Vane et al., 2011). Anthropogenic Pb emissions are thus a potentially valuable source of chronological control on salt-marsh accretion history over the last 200 years.

Changes in measured Pb isotopes caused by emissions from vehicles using leaded gasoline can be verified by concentrations of antimony (Sb), because it is a high-contrast marker for aerial deposition from road traffic (Gomez et al., 2005; Amarasiriwardena and Wu, 2011; Fujiwara et al., 2011). Three elements (Zn, Sb & Br) have particular utility as motor vehicle trace element markers because they occur in fine and coarse particulates (Huang et al., 1994). Of these, Sb has the lowest natural low abundance in pristine environments with concentration in the upper crust estimated to be 0.31 mg/kg (Wedepohl, 1995). Naturally elevated concentrations of Sb can be associated with mineralization. Smelting of these mineralized ores will have generated aerosol deposition and elevated background levels since industrialization (Gal et al., 2007). Sb has similar environmental behavior to Pb (Shotyk et al., 2005) and its potential for long distance transport (and subsequent deposition in salt-marsh sediments) from traffic emissions has been demonstrated by its enrichment in the PM10 fraction (Amato et al., 2009).

In this paper we investigate Pb and Sb concentrations and Pb isotopes (^{210}Pb with ^{137}Cs and ^{206}Pb:^{207}Pb) as a chronometer of recent salt-marsh sediments using a sediment core from a U.S. mid-Atlantic salt marsh.

2. Regional setting

Sediment core BB1 was obtained from a salt marsh in northern Barnegat Bay (New Jersey), an archetype back-barrier coastal lagoon in the U.S. mid-Atlantic region (Fig. 1). The 280 km^2 bay, which is fringed by *Spartina alterniflora* salt marsh, has a mean depth of 1.5 m and a mean tidal range that decreases from 1.4 m at the inlet to 0.3 m at its northern and southern limits. Although the bay and its extensive marshes have a history of human impacts, including grid ditching (Kennish, 2001) and eutrophication (Kennish et al., 2007), much of the marsh accretes vertically unobstructed by human influences. Barnegat Bay lacks a large source of river sediment, therefore supply of allochthonous mineral sediment to the marsh platform is low compared to mid-Atlantic river-estuarine marshes. Consequently, salt-marsh accretion rates determined by ^{210}Pb and ^{137}Cs measurements at sites throughout the bay are low (0.2–0.3 cm/yr) compared to minerogenic marshes in the region (Velinsky et al., 2011). Importantly, supply of ^{210}Pb to
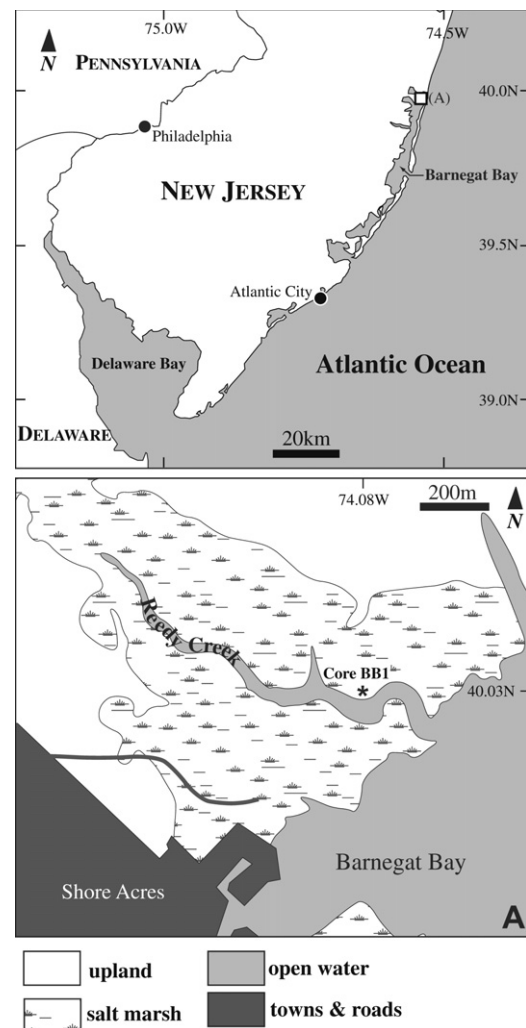

Fig. 1. Location of the Barnegat Bay study site in New Jersey, USA. Core BB1 has the geographic coordinates 40.02988° N, 74.07995°W.

Barnegat Bay salt marshes is derived mostly (if not exclusively) from direct atmospheric deposition, an ideal condition for developing chronologies with anthropogenic Pb isotopes. Core BB1 was selected because it was representative of the stratigraphy at the study site, exhibited concordant ^{210}Pb and ^{137}Cs chronologies, and was sufficiently long (94 cm) to capture the full synindustrial record of anthropogenic Pb fallout.

3. Methods

Sediment cores from Barnegat Bay salt marshes were collected in 2009 as part of an unrelated study of historical sediment and nutrient loading (Velinsky et al., 2011). Using a push-piston core (10 cm diameter) system designed to minimize compaction, BB1 was collected in a high-marsh floral zone (short-form *S. alterniflora*), 25 m landward of Reedy Creek, at a surveyed elevation of 0.10 m NAVD 88 (Fig. 1). Duplicate cores of BB1 were recovered, one for radionuclide and chemical analysis and another for stratigraphic description and archiving. Cores were capped in the field and transported to the laboratory for processing.

3.1. Sample preparation

The core for radionuclide and chemical analysis was sectioned in 2 cm thick intervals. Samples were weighed wet, dried at 110 °C for 24 h in a convection oven, and weighed again to determine gravimetric water content, porosity, and dry-bulk density (Bennett and Lambert, 1971). Approximately 20–60 g of dried sediment was ground to a homogeneous powder, and a 4 g aliquot was combusted in a muffle furnace to determine loss on ignition (LOI) following methods described in (Heiri et al., 2001). LOI quantified the relative proportion of organic (combustible) and mineral (residual ash) materials in the sediments. Gravimetric and LOI data were used to interpret the downcore radionuclide and stable Pb profiles. Each sample between depths of 0 and 94 cm ($n = 47$) was analyzed for Pb concentrations and stable Pb isotopes, whilst only samples from 0 to 36 cm ($n = 18$) were analyzed for ^{210}Pb and ^{137}Cs activity.

3.2. Radionuclide measurement

Powder samples were sealed in a 60 ml plastic jar, stored for at least 30 days to ensure equilibrium between [226]Ra and [214]Bi, and then counted for 24 h on Canberra Model 2020 low-energy Germanium detectors. Measurements of [210]Pb ($t_{1/2} = 22.3$ years) and [137]Cs ($t_{1/2} = 30.1$ years) activity were made by gamma spectroscopy of the 46.5 and 661.6 keV photopeaks, respectively (Cutshall et al., 1983; Wallbrink et al., 2002). The full-energy peak efficiency of these detectors is 4% at 46.5 keV and 1% at 661.6 keV, and the minimum detectable activity for photopeaks of ^{210}Pb and ^{137}Cs is approximately 2–3 Bq/kg. Excess ^{210}Pb activity was determined by subtracting the activity of its parent nuclide ([214]Bi at 609.3 keV) from the total activity ($^{210}Pb_{xs} = {}^{210}Pb_{tot} - {}^{214}Bi$). Detector efficiencies were determined using the NIST Ocean Sediment Standard Reference Material 4357 (Inn et al., 2001). The standard (a sediment powder) and core material were counted in an identical geometry, thus negating a self-absorption correction for [210]Pb. Confidence limits for radionuclide data are computed as the propagated one-sigma background, calibration, and counting errors.

3.3. Radionuclide chronology

Two models were used to develop ^{210}Pb chronologies for core BB1. Detailed descriptions of both approaches are available in the literature (Robbins, 1978; Appleby and Oldfield, 1992; Appleby, 2001). The Constant Initial Concentration (CIC) model assumes that the *specific activity* (Bq/g) of excess ^{210}Pb deposited on a marsh surface remained constant through time and that variations in the rate of sediment deposition do not influence the initial activity of excess [210]Pb. The Constant Rate of Supply (CRS) ^{210}Pb model relates sediment age and depth by assuming that the *depositional flux* of [210]Pb (Bq/cm^2/yr) to a marsh surface was constant. Peak ^{137}Cs at AD 1963–64 provided an independent means to corroborate ^{210}Pb accumulation histories.

3.4. Determination of Pb and Sb concentrations and ratios of stable Pb isotopes

In preparation for determining concentrations of stable Pb isotopes and Sb, 0.25 g of powdered sediment was dissolved by an HF/HClO$_4$/HNO$_3$ mixed concentrated acid attack in Savillex™ PFA vials. Samples were reconstituted in dilute nitric acid and diluted to: (i) within the calibration range of Pb and Sb chemical standards for concentration measurements; and (ii) within the pulse counting range (<1Mcps) of the ICP-MS for isotope ratio measurements.

Concentration and isotope ratio determinations were made using a quadrupole ICP-MS instrument (Agilent 7500c) with a conventional glass concentric nebulizer. The long term 2σ precision for the BCR-2 reference material used for quality control, which has a total Pb concentration of 11 mg/kg was $^{207/206}Pb = 0.0008$, $^{208/206}Pb = 0.0020$, based on $n = 32$ replicates over 29 months and a mean accuracy, relative to the published values of (Baker et al., 2004), within that error. Data (raw isotope intensity count rates) were processed off-line using Microsoft Excel spreadsheets. Processing consisted of: (i) removal of background; (ii) calculation of isotope ratios; (iii) determination of mass bias correction factor from defined isotope ratios of SRM981; (iv) application of mass bias factors derived from SRM981 using external standard-sample-standard bracketing; and (v) optional further correction for linearity of ratio with signal strength.

3.5. Identifying Pb chronostratigraphic markers

We identified nine historical features of U.S. Pb production and consumption that could be used as chronostratigraphic markers to estimate sediment age in core BB1 (Table 1; Fig. 2). Four were recognized in Pb concentration (A–D) and five in [206]Pb:[207]Pb ratios (E–I). Changes in production and consumption were assumed to have caused a corresponding change in Pb emissions that were transported through the atmosphere by constant prevailing wind patterns and deposited on the salt-marsh surface within a few years (Graney et al., 1995) and without isotopic fractionation (Ault et al., 1970). As emissions per unit of production or consumption are likely to have changed over time, trends rather than absolute values are the basis for recognizing these features in core BB1. The stratigraphic context of core BB1, and the ordering of samples, provides an appropriate sedimentary constraint for interpreting downcore changes in Pb concentration and [206]Pb:[207]Pb as being reliably correlated to the nine historical features. We used downcore concentration of Sb to help distinguish between changes before and after the widespread introduction of automobiles around AD 1920 because concentrations raised above low background levels are frequently caused by motor vehicles (Gomez et al., 2005; Amarasiriwardena and Wu, 2011; Fujiwara et al., 2011). This association with automotive emissions makes it useful for confirming interpretations of Pb data as being related (or unrelated) to leaded gasoline.

Table 1

Ages assigned to samples in core BB1 using measured Pb concentrations and ^{206}Pb:^{207}Pb ratios in comparison with historical U.S. and Upper Mississippi Valley (UMV) Pb production and consumption. Age error is an estimate of the uncertainty of identifying a specific date in historical records and is a minimum of 5 years to include a lag between Pb emission and deposition. Depth has an uncertainty for sample thickness (± 1 cm) and in some instances is larger because age horizons could be associated with multiple, adjacent samples.

	Year	Type	Description	Depth in BB1 (cm)	Age error ($\pm$years)
A	1875	Pb concentration	Start of national Pb production	33 ± 3	5
B	1925	Pb concentration	Peak in national Pb production	21 ± 1	5
C	1935	Pb concentration	Minimum in national Pb production and consumption	17 ± 1	6
D	1974	Pb concentration	Peak in national Pb production	9 ± 1	5
E	1827	^{206}Pb:^{207}Pb	Start of UMV Pb production	41 ± 1	5
F	1857.5	^{206}Pb:^{207}Pb	Peak contribution by UMV	34 ± 2	5
G	1880	^{206}Pb:^{207}Pb	Decline of UMV Pb production	25 ± 3	20
H	1965	^{206}Pb:^{207}Pb	Gasoline minimum	11 ± 1	5
I	1980	^{206}Pb:^{207}Pb	Gasoline peak	7 ± 1	5

4. Results

4.1. ^{210}Pb and ^{137}Cs activities and chronology

Radionuclides were measured in the upper 36 cm of core BB1 (Fig. 3a). Excess ^{210}Pb increased near-exponentially upcore from 1.3 Bq/kg at 31 cm to 166 Bq/kg at the top, a pattern consistent with steady-state sediment accumulation and radioactive decay. The accretion rate based on regression of the decay profile is 0.25 cm/yr. The ^{137}Cs profile (Fig. 3b) broadly mirrors the record of ^{137}Cs atmospheric fallout in northern temperate latitudes (Warneke et al., 2002). Activity of ^{137}Cs increased upcore from the depth of first occurrence above the detection limit (20—22 cm) to a peak centered at 11 cm, above which activity decreased to the core top. The accretion rate based on the AD 1963—64 peak is 0.25 cm/yr, identical to the ^{210}Pb-based rate. Accumulation histories were developed for BB1 using the CIC and CRS models (Fig. 3c). This yielded dates from AD 1901 to the time of core collection (AD, 2009).

4.2. Total Pb and Sb concentration and ratios of stable Pb isotopes

The average Pb concentration of samples between 93 cm and 43 cm in BB1 was 11.2 mg/kg and represents background levels (Fig. 4c). Samples at 41, 39 and 37 cm had an average Pb concentration of 23.8 mg/kg. From 35 cm to 21 cm, it increased from 20.5 to 93.3 mg/kg. The interval from 21 cm to 17 cm was characterized by a decline in Pb concentration to 76.5 mg/kg. Maximum Pb concentration (164 mg/kg) occurred at 9 cm and declined to 65.8 mg/kg at the modern marsh surface. Antimony (Sb) concentrations paralleled those of Pb (Fig. 4c). Below 41 cm, total Sb concentration was 0.09—0.36 mg/kg. It increased to a peak of 1.44 mg/kg at 23 cm before declining to 1.13 mg/kg at 13 cm. Maximum Sb concentration was at 9 cm (1.63 mg/kg) and declined to 0.86 mg/kg at the core top.

Measured concentrations of Pb isotopes are expressed as ratios. From 93 cm to 51 cm ^{206}Pb:^{207}Pb varied between 1.197 and 1.226 (Fig. 4d). It reached a minimum of 1.174 at 43 cm before increasing to a maximum value of ~1.234 at 33—35 cm. There was a decline in ^{206}Pb:^{207}Pb between 33 cm and 11 cm (1.186) prior to another increase to 1.207 at 9 cm and a subsequent decrease to the modern marsh surface value of 1.200.

5. Discussion

5.1. ^{210}Pb and ^{137}Cs accumulation histories

The CRS model, which was originally developed for lake sediments (Krishnaswamy et al., 1971) and only later applied to salt marshes (McCaffery and Thomson, 1980), is best suited to marshes that sequester ^{210}Pb primarily from direct atmospheric deposition. In highly allochthonous marsh systems, particle-borne^{210}Pb transported tidally to the marsh platform supplies activity in addition to that derived atmospherically. Because flux of tidal ^{210}Pb to the sediment surface is highly discontinuous, the steady-state assumption of the CRS model is frequently contradicted in minerogenic marshes. Measured inventories of excess ^{210}Pb and ^{137}C in Barnegat Bay salt-marsh sediments are comparable to theoretical inventories supported by the regional atmospheric flux, suggesting that tidal supply of these radionuclides is minimal (Velinsky et al., 2011). Indeed, excess ^{210}Pb and ^{137}Cs inventories at Site BB1 are 394 ± 3.5 mBq/cm^2 and 116 ± 9.3 mBq/cm^2, respectively, and fall within the range of reference inventories (327—523 mBq/cm^2 for excess ^{210}Pb and 82—250 mBq/cm^2 for ^{137}Cs) reported for the U.S. Atlantic coast (Graustein and Turekian, 1986; Todd et al., 1989).

5.2. Pb concentrations

Four features of historic U.S. Pb production and consumption (Fig. 2a) are recognized in BB1 (Fig. 4c; Table 1). We assumed that Pb deposited in Barnegat Bay during the 20th century was transported by unchanged prevailing winds from industrial centers in the U.S, that this continental-scale signal overwhelmed any local sources and that the U.S. dominated (or was at least representative of) North American Pb production. Prior to AD 1935, U.S. consumption of Pb was equal to primary production. Pb production and consumption increased dramatically in the second half of the 19th century, although some anthropogenic Pb was released to the atmosphere much earlier in North America (Heyl et al., 1959) and Europe (Rosman et al., 1997). During this period inefficient furnaces and smelting coupled with development of taller stacks caused large amounts of Pb to be released to the atmosphere (Nriagu, 1998). National Pb production increased from an annual average of 15,703 tons between AD 1830 and AD 1871 to 106,218 tons in AD 1881 (Fig. 2a). The pronounced increase in Pb concentration at 33 ± 3 cm in BB1 represents this onset of large-scale, national Pb production and consumption. To accommodate uncertainty in establishing the timing of this change we assigned the sample a date of AD 1875 $\pm$ 5 years. The vertical uncertainty reflects difficulty in attributing the start of a trend to a specific sample (those at 35, 33 or 31 cm could represent this event). U.S. Pb production peaked in AD 1925 (620,913 tons) and consumption peaked in AD 1928, which is recorded at 21 cm in BB1 with an estimated uncertainty of ± 5 years.

The period from AD 1933 to AD 1962 was characterized by lower Pb production (average 319,244 tons annually). Coincident with the Great Depression, Pb consumption declined from 683,655 tons in AD 1930 to 378,024 in AD 1932 (Fig. 2a). However, it had recovered to 952,544 tons in AD 1941. This divergence between production
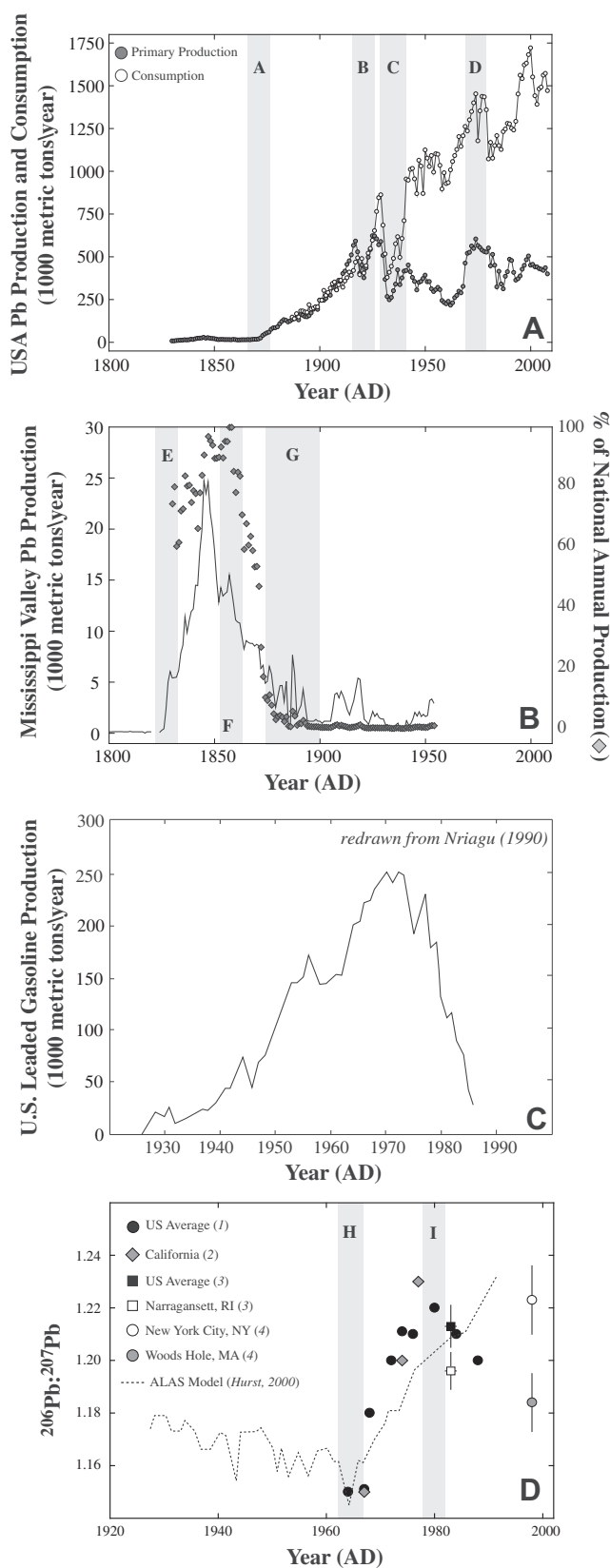
Fig. 2. Historical records of Pb production and consumption. Vertical grey bars labeled A–I correspond to historic features used to identify chronostratigraphic markers in core BB1 (A) U.S. Pb primary production and consumption. Data from the USGS Lead Statistical Compendium (AD 1830 to AD 1990) and the USGS Minerals Yearbook (AD 1991 to AD 2008). (B) Pb production from Upper Mississippi Valley ores (solid line from Heyl et al., 1959), also shown as a percentage of the U.S. total (grey diamonds,

and consumption was a consequence of increasing consumption of secondary and imported Pb. A minimum Pb concentration at 17 cm in BB1 was interpreted as corresponding to the consumption decline and assigned a date of AD 1935 ± 6 years. A second peak in Pb production (602,253 tons) and consumption (1,450,976 tons) occurred in AD 1974, since when it followed a declining trend. Peak Pb concentration in BB1 at 9 cm was assigned a date of AD 1974 ± 5 years.

5.3. Pre-AD 1920 stable Pb isotopes

Prior to the 1920s, the principal source of anthropogenic Pb released to the atmosphere was industrial activity. Between AD 1830 and AD 1870 most U.S. Pb (average 79% ± 17%, 1σ) was sourced from the Upper Mississippi Valley (UMV) Pb and Zn district (Fig. 2b), principally in Illinois, Iowa and Wisconsin where it was smelted close to mines (Heyl et al., 1959). UMV ores are distinctive because of their unusual isotopic composition with high (1.3–1.5) ^{206}Pb:^{207}Pb values (Heyl et al., 1966, 1974; Doe and Delevaux, 1972). Three features of historic UMV Pb production are recognized in BB1 (Fig. 4d; Table 1). Ratios of ^{206}Pb:^{207}Pb measured in BB1 likely record changes in UMV Pb production because of the location of Barnegat Bay relative to this source and prevailing winds that carried UMV Pb to regions north and south of New Jersey (Marcantonio et al., 2002; Lima et al., 2005a) as well as to Bermuda (Kelly et al., 2009). Therefore ^{206}Pb:^{207}Pb measurements in New Jersey salt-marsh sediments can potentially be used as chronostratigraphic markers if the signal from UMV production is assumed to be dominant over local sources that were small until the late 19th century (Bleiwas and DiFrancesco, 2010). Dates ascribed to depths in core BB1 using Pb concentrations are stratigraphic constraints on the timing of ^{206}Pb:^{207}Pb changes (Fig. 4). Samples below 43 cm show variability, but do not represent anthropogenic Pb inputs because these samples have background Pb concentrations. Sb concentrations confirm that changes associated with UMV Pb production took place before the widespread use of automobiles.

UMV Pb production increased rapidly between AD 1824 (301 metric tons) and AD 1830 (5416 metric tons; Heyl et al., 1959). We assigned the initial increase in measured ^{206}Pb:^{207}Pb at 41 cm a date of AD 1827 ± 5 years to correspond with the start of UMV Pb production. Peak production (approximately 25,000 metric tons annually) occurred from AD 1845 to AD 1847 (Heyl et al., 1959). Peak ^{206}Pb:^{207}Pb in BB1 was at 34 cm (±2 cm). Measured ^{206}Pb:^{207}Pb peaks in sediment may better reflect maximum proportional contribution than maximum absolute production (in tons) because this is the time when dilution of the UMV signal from other sources was minimal. UMV Pb made its maximum relative contribution to national production in AD 1857 and AD 1858. Therefore the peak at 34 cm was assigned a date of AD 1857 ± 5 years.

Production of Pb in the UMV declined during the late 19th century from 47% of national output in AD 1871 to less than 5% by AD 1878. It had fallen to AD 1827 levels by the middle of the AD 1880s and from AD 1895–1905 average production was 180 tons annually equating to <1% of national production. We assigned a date of AD 1880 ± 20 years to this decline. Identifying its effect on measured ^{206}Pb:^{207}Pb in BB1 is challenging; a minimum occurred at 21 cm although the decline to background values

right axis). (C) Annual production of leaded gasoline in the U.S., redrawn from Nriagu (1990). (D) Historic record of ^{206}Pb:^{207}Pb in US leaded gasoline and atmospheric measurements. The Anthropogenic Pb ArchaeoStratigraphy (ALAS) model (dashed line) is reproduced from Hurst (2000). Data points are atmospheric measurements of ^{206}Pb:^{207}Pb from locations in the USA (and in particular the northeast) reported by (1) Rosman et al. (1993) (2) Shirahata et al. (1980) (3) Sturges and Barrie (1987) and (4) Bollhöfer and Rosman (2001).
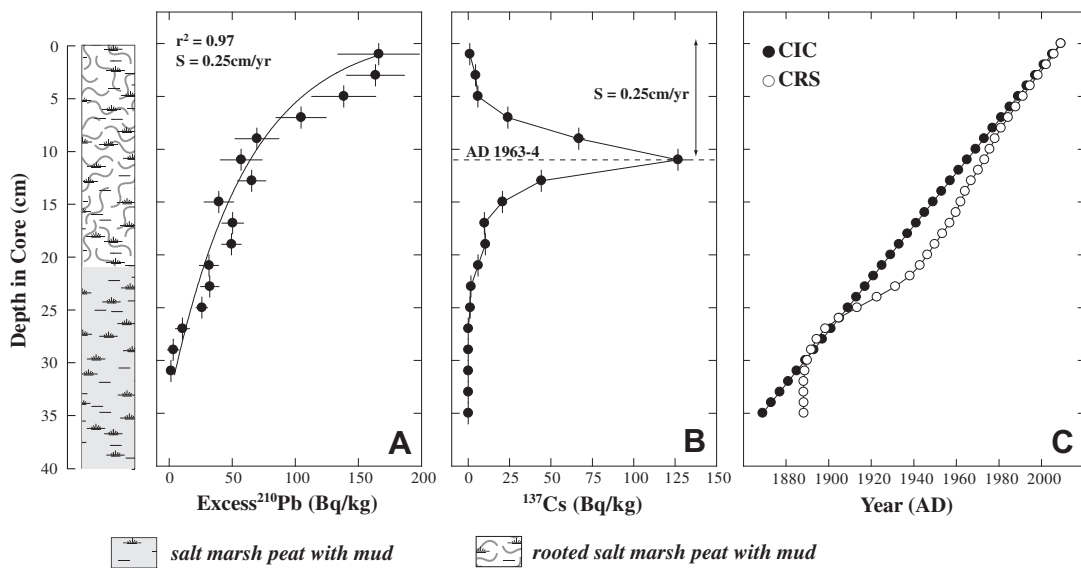
Fig. 3. Activity profiles of ^{210}Pb and ^{137}Cs in core BB1. (A) Profile of excess ^{210}Pb with a fitted exponential curve and the accretion rate (S). Analytical uncertainty and sample thickness are shown by error bars for each point. (B) Profile of ^{137}Cs showing an activity peak assumed to be concordant with AD 1963−1964, and the accretion rate (S). Analytical uncertainty is smaller than symbol. (C) Age-depth models developed for core BB1 using the ^{210}Pb CIC and CRS models.

(approximately 1.207) occurred at 26 cm. This is further complicated by rapidly increasing national production that masks and overprints changes in the UMV. We assigned the sample at 23 cm a date of AD 1880 ± 20 years with an added vertical range of ±3 cm to capture the uncertainty described. Disassociation of these changes with Sb concentrations suggests that they were not caused by emissions from automotive transport and occurred prior to AD 1920. Although absolute production of UMV Pb increased during the 20th century (e.g. World War One) it did not again exceed 1.5% of the national total.

Following isotopic analysis of other potential sources including coal and Pb from other regions, Lima et al. (2005a) concluded that

downcore [206]Pb:[207]Pb changes corresponded to historical Pb production in the UMV. Marcantonio et al. (2002) showed that Chesapeake Bay estuarine sediments deposited in the middle of the 19th century had a broad, shallow [206]Pb:[207]Pb peak (Fig. 5a). Great Lake sediments dated using ^{210}Pb and pollen had peak anthropogenic [206]Pb:[207]Pb at AD 1883 in Lake Erie, AD 1863 in Lake Michigan and AD 1895 in Lake Ontario (Graney et al., 1995). In Florida, a core of peat dated using ^{210}Pb and ^{14}C did not show significant increase in [206]Pb:[207]Pb during the 19th century (Kamenov et al., 2009). Annually-banded corals provide an independently dated archive of historical Pb deposition (Shen and Boyle, 1987). At John Smith's Bay (Bermuda), [206]Pb:[207]Pb increased from 1.187 in AD 1826 to 1.211 in
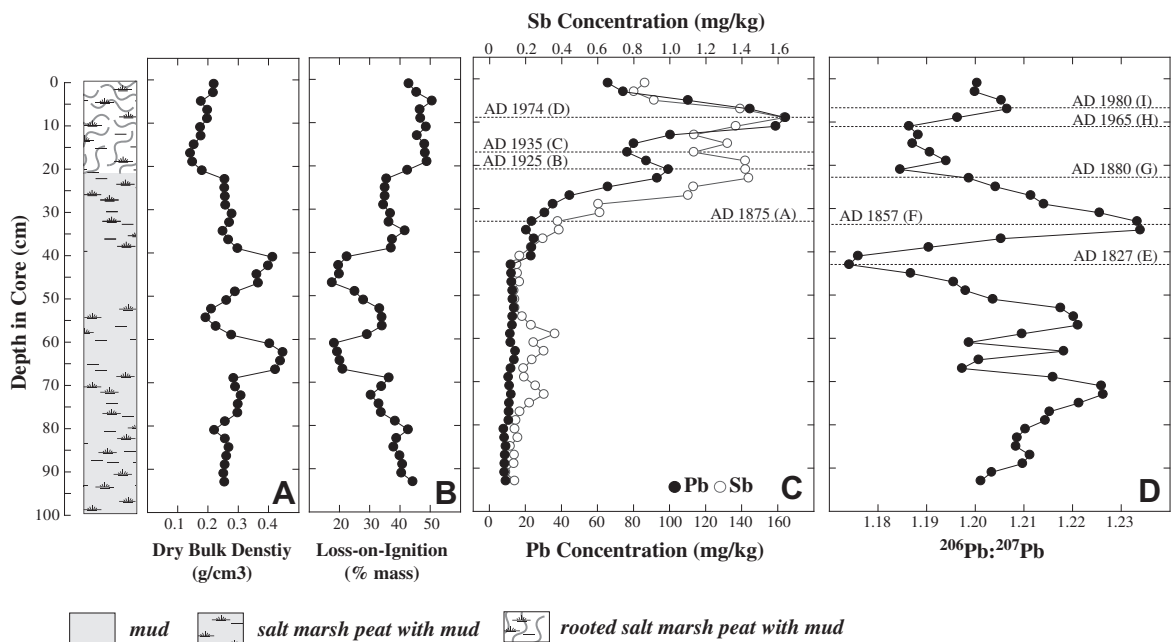

Fig. 4. Downcore measurements of stable Pb concentrations in Core BB1. (A) Gravimetric dry-bulk density. (B) Organic matter mass content estimated by loss-on-ignition (LOI). (C) Concentration of Pb (closed circles, black line) and Sb (open circles, grey line). (D) Measured ratio of [206]Pb to [207]Pb. Labeled dashed lines mark features used in assigning dates to samples in core BB1.
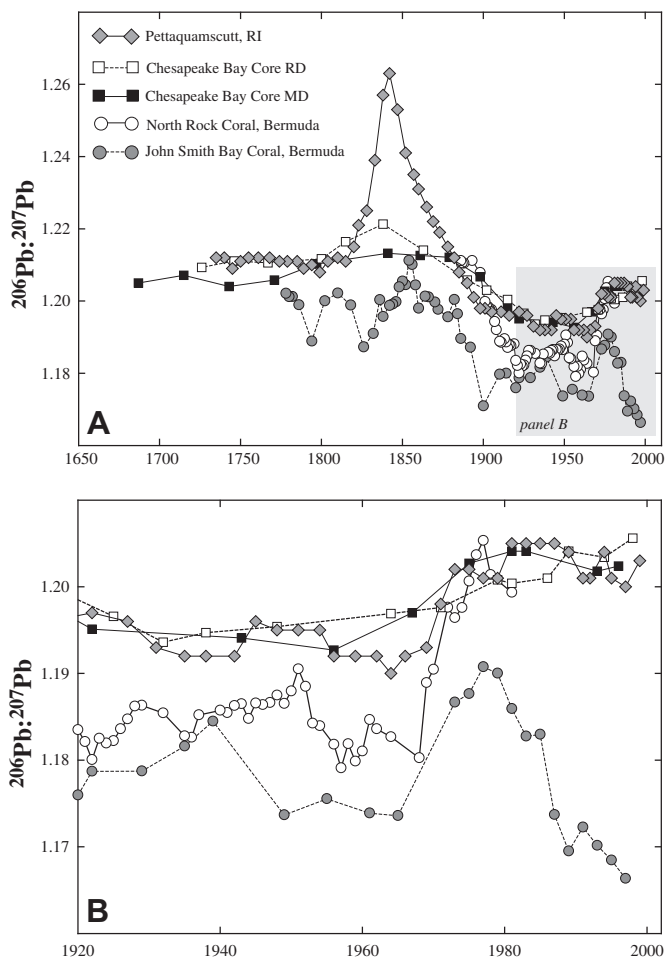
Fig. 5. Sedimentary records of ^{206}Pb:^{207}Pb changes through time. (A, B) Historic fluctuations in ^{206}Pb:^{207}Pb preserved in river basin (Pettaquamscutt, RI; Lima et al., 2005a) and estuarine (Chesapeake Bay; Marcantonio et al., 2002) sediment cores and in annually-banded corals (Bermuda; Kelly et al., 2009). Chronological control in each case was from an independent dating method such as varve or band counting, ^{210}Pb or pollen changes of known age. Changes in ^{206}Pb:^{207}Pb were not used as a dating technique.

AD 1854, which was attributed to UMV Pb production (Kelly et al., 2009). In Rhode Island, Lima et al. (2005a,b) identified an increase in measured ^{206}Pb:^{207}Pb from 1.211 (AD, 1815) to 1.325 (AD, 1842) in lake sediments dated using varve counting confirmed by ^{210}Pb with ^{137}Cs peaks (Fig. 5a). They concluded that differences among timing of mid-19th century ^{206}Pb:^{207}Pb maximums are caused by problems with age models and that the peaks actually represent simultaneous deposition of UMV Pb. Differences in the magnitude of ^{206}Pb:^{207}Pb peaks were proposed to reflect mixing of multiple, spatially variable Pb sources (Lima et al., 2005a). For example, prevailing winds caused less UMV Pb to be deposited in the Chesapeake Bay than in New England and its absence in Florida. Consequently, Lima et al. (2005a) proposed that three chronological horizons (initiation, peak and decline) associated with deposition of UMV Pb could be identified in sedimentary archives in the northeastern U.S. These horizons narrow the chronological hiatus between radiocarbon and ^{210}Pb accumulation histories.

5.4. Post-AD 1920 stable Pb isotopes

Two ^{206}Pb:^{207}Pb features associated with leaded gasoline are recognized in BB1. Leaded gasoline was a significant source of anthropogenic Pb following its introduction in AD 1923 (Facchetti, 1989; Nriagu, 1990). Peak U.S. production occurred in AD 1970 (exceeding 250,000 tons annually; Fig. 2c). Following the Clean Air Act (AD 1970) consumption of leaded gasoline declined to 17,000 tons in AD 1988 (Nriagu, 1990). By AD 1993 U.S. Pb emissions from gasoline were 1% of those in AD 1970 (Bollhöfer and Rosman, 2001). Up to two thirds of Pb added to gasoline entered the atmosphere as fine particulate matter that was transported long distances through the atmosphere (Facchetti, 1989). The Pb ore(s) used in gasoline production and released to the atmosphere varied over time and also among regions in the U.S. depending on the mixture used by each manufacturer (Shirahata et al., 1980; Facchetti, 1989; Rosman et al., 1993; Graney et al., 1995), although geological records of Pb deposition and direct measurements of atmospheric Pb isotopes suggest that a regional-scale patterns can be discerned for the northeastern U.S. (Marcantonio et al., 2002).

Direct measurements of the isotopic composition of gasoline or atmospheric emissions are sparse prior to about AD 1965. The ALAS model (Hurst, 2000) suggests that ^{206}Pb:^{207}Pb in gasoline declined from 1.175 in the 1920s to 1.145 in the mid-1960s (Fig. 2d). These values are in agreement with atmospheric measurements for eastern North America of 1.15 in AD 1967 (Shirahata et al., 1980; Rosman et al., 1993). In Rhode Island lake sediments, Lima at al. (2005a) reported a decline from 1.197 in AD 1922 to 1.190 in AD 1964 (Fig. 5b). Similarly, sediments from the Chesapeake Bay (core MD) showed a ^{206}Pb:^{207}Pb decline from 1.207 in AD 1898 to a minimum of 1.193 in AD 1956. Coral records from Bermuda showed no trend in isotopic ratio (Kelly et al., 2009). Decreased ^{206}Pb:^{207}Pb from 19 cm to 11 cm in BB1 (Fig. 4d) was caused by the introduction (and dominance until the mid 1960s) of leaded gasoline with a relative constantly isotopic signature of 1.165 (Hurst, 2000; Lima et al., 2005a). This had the effect of diluting other contributions and caused the minimum of 1.185 at 11 cm in BB1. We assigned this ^{206}Pb:^{207}Pb horizon a date of AD 1965 ± 5 years (Table 1; Fig. 4d).

After approximately AD 1965, ^{206}Pb:^{207}Pb ratios in gasoline increased (Fig. 2d) because Pb ores from Missouri were increasingly used (Graney et al., 1995; Hurst, 2000). Shirahata et al. (1980) showed an increase in measured atmospheric ^{206}Pb:^{207}Pb from 1.15 in AD 1967 to 1.23 in AD 1977, whilst the averaged record of Rosman et al. (1993) showed a U.S. aerosol peak of 1.22 in AD 1980. Peak ^{206}Pb:^{207}Pb in Greenland snow occurred between AD 1972 and AD 1984 (Rosman et al., 1993). Since the decline of leaded gasoline, no clear regional pattern of changing ^{206}Pb:^{207}Pb values has emerged and records show variability since AD 1980 (Fig. 2d). The ^{206}Pb:^{207}Pb peak in BB1 at 7 cm was given a date of AD 1980 ± 5 years (Table 1; Fig. 4d). The sedimentary record from Rhode Island showed a ^{206}Pb:^{207}Pb rise from 1.192 in AD 1966 to a peak of 1.205 in AD 1981 (Lima et al., 2005a). In the Chesapeake Bay, ^{206}Pb:^{207}Pb values increased from 1.197 in AD 1967 to a peak of 1.204 in AD 1981. Bermudan corals show a pronounced increase in ^{206}Pb:^{207}Pb between AD 1964 (1.174 and 1.183 at John Smith's Bay and North Rock respectively) and AD 1977 (1.191 and 1.205).

5.5. Development of age-depth profiles for BB1

We used the nine samples with ages estimated from Pb concentrations or ^{206}Pb:^{207}Pb (Table 1) as the input for an age-depth model (Fig. 6). In addition, year of core collection (AD, 2009) was added as a surface data point (0–1 cm depth), but was not specified as a fixed constraint. Results from ^{210}Pb and ^{137}Cs were not included. Age-depth models use dated samples to provide downcore age estimates with sample-specific uncertainty at 1 cm intervals. We applied the "Clam" model of Blaauw (2010) executed in R and using a locally-weighted spline. Model age uncertainties (2σ) ranged from 4 to 15 years with an average of 8 years. The Clam model estimates an approximately linear rate of sediment
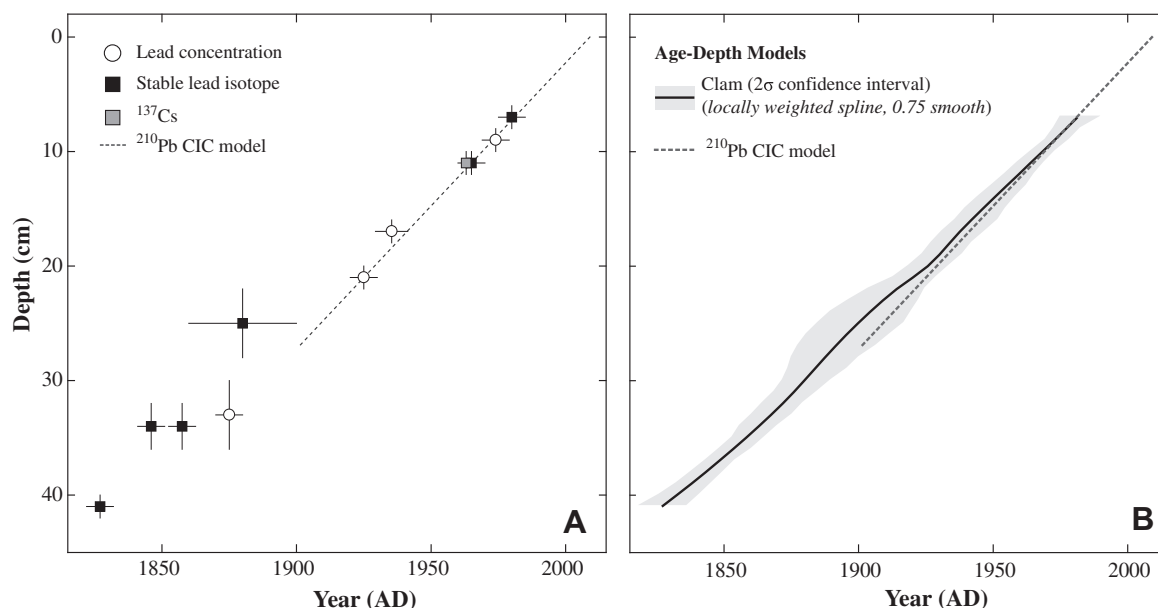
Fig. 6. Age-depth estimates for core BB-1. (A) Downcore changes in Pb concentration (open circles) and ^{206}Pb:^{207}Pb ratios (filled squares) were used to assign dates to specific depths on the basis of documented changes in national Pb production and consumption and the contribution of Upper Mississippi Valley Pb with its unusual isotopic signature. Isotopic changes at 34 cm could reflect maximum absolute production in the Upper Mississippi Valley (AD 1846) or maximum proportional contribution (AD 1857), both are shown for comparison. Dashed grey line shows age-depth relationship from a constant initial concentration (CIC) model of ^{210}Pb accumulation that is supported by peak ^{137}Cs activity (grey square). It is restricted to the upper 27 cm of the core because at lower depths excess ^{210}Pb did not exceed analytical error. Vertical error bars represent sample thickness and uncertainty in selecting a single sample to assign an age to. Horizontal error bars represent uncertainty in assigning features in the historic record of Pb production an age, in most cases ± 5 years was used. (B) Age-depth model produced by "Clam" (Blaauw, 2010) using the nine horizons from changes in Pb concentration and ^{206}Pb:^{207}Pb ratio. A date of AD 1857 was used at 34 cm. The model was developed a locally-weighted spline function. Grey error band is the 95% confidence interval for estimated ages (solid line). CIC accumulation model developed from ^{210}Pb and ^{137}Cs is shown as a dashed grey line.

accumulation in core BB-1 since AD 1827 (lowest date), although a period of slightly reduced sedimentation is predicted between approximately AD 1910 and AD 1940 (Fig. 6b). The age-depth model developed using CIC ^{210}Pb accumulation lies within the 95% confidence interval of the Clam model at all depths. Although both techniques use Pb deposited from the atmosphere, they have different origins and are independent of one another. Their agreement suggests that age-depth relationships developed using ^{210}Pb accumulation and horizons of Pb concentration and ^{206}Pb:^{207}Pb provided chronologies that are in agreement and serve to mutually reinforce the validity of both approaches.

6. Conclusions

Anthropogenic Pb emissions preserved in salt-marsh sediment can be used as chronological markers to reconstruct environmental change and wetland development prior to the instrumental period. Changing downcore Pb concentrations from a salt marsh in New Jersey, USA reflected prominent features in historic U.S. production and consumption at AD 1875, 1925, 1935 and 1974. Stable Pb isotopes transported by prevailing winds correlated with production in the Upper Mississippi Valley where Pb ores have an unusual and distinctively high ^{206}Pb:^{207}Pb ratio (AD, 1827, 1857 and 1880). Changes in leaded gasoline are recognized at AD 1965 and 1980. These markers meet the need for geochronological horizons in the radiocarbon plateau of the last 350 years and also extend and independently corroborate accumulation histories derived from ^{210}Pb for the last ∼ 100 years.

Acknowledgments

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